Resonance structure of composite and slightly absorbing spheres

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Using optical levitation of individual 3–10-μm droplets, we investigate the resonance structure of the light intensity scattered at an angle of approximately 90° by a dielectric droplet, a slightly absorbing droplet, and composite inhomogeneous droplets. We find in all the cases that the resonance structures of dielectric, weakly absorbing, and composite inhomogeneous spheres are almost identical for the imaginary part of the refractive index (or of an effective refractive index for the case of a composite sphere) smaller than 10⁻⁵. Thus a small amount of highly absorbing material dispersed in a dielectric sphere does not destroy its resonance characteristics.

INTRODUCTION

The optical or the electromagnetic levitation of homogeneous spherical droplets and the observation of the Mie scattering resonance structure in the radiation pressure or in the scattered light intensity stimulated a period of intense theoretical and experimental research in the field of the interaction of electromagnetic radiation with micrometer-sized droplets. Most investigations in the field have been limited to dielectric and homogeneous materials. For a homogeneous sphere the scattered and the inside electromagnetic fields can be readily calculated by using the Mie theory, and thus the comparison between the experimental results and the theory is readily available.

Calculation suggests that the electromagnetic energy density varies by several orders of magnitude within a spherical droplet, depending on the location, the size parameter, and the refractive index of the droplet. The maximum values of electromagnetic fields are usually located at the surface or close to the surface of a droplet. The field enhancement that is due to the droplet can be quantitatively characterized by the source function \( S = |E(r)/E_0|^2 \), where \( E(r) \) is an electromagnetic-field vector at the investigated point and \( E_0 \) is an electromagnetic-field vector of the incident radiation. For an arbitrarily chosen size parameter and refractive index of a spherical particle, the typical values of the maximum of the source function are in the range 10–100. However, whenever definite resonance conditions are satisfied, the source function reaches values for (experimentally observable resonances) of the order of 10⁴–10⁶. The role of the field enhancement that is due to a homogeneous spherical particle has been clearly demonstrated in the case of stimulated Raman scattering, the laser-induced breakdown of gases, and other phenomena.

A question of considerable practical interest concerns whether an inhomogeneous sphere composed of a small amount of highly absorbing material embedded in a dielectric medium will have a similar set of resonances and internal field structure as a same-size homogeneous dielectric sphere. The research reported here is focused on the experimental investigation of the resonances of inhomogeneous spheres.

RESONANCES OF SPHERICAL PARTICLES

Resonances in the Mie scattering amplitudes \( a_n \) and \( b_n \) are responsible for the ripple structure of the extinction, the scattering, and the absorption efficiencies \( Q_{\text{ext}}, Q_{\text{s}}, \) and \( Q_{\text{abs}} \), respectively, as well as for sharp peaks in all the other scattering characteristics including the differential scattering cross section. The resonances are usually treated as poles in the amplitudes \( a_n \) and \( b_n \) and are manifested as extremes in \( \text{Re}(a_n) \) and \( \text{Re}(b_n) \) or in \( |a_n| \) and \( |b_n| \).

Since the imaginary part of the partial-wave amplitude changes sign and passes through zero from a positive to negative values (with the increasing value of the size parameter \( x \)) at the resonance position, it is possible to identify the resonance position with this zero value of the imaginary part. Thus some computer programs locating resonance positions are based on the zero-crossing properties of an imaginary part of \( a_n \) or \( b_n \).

The case of the complex refractive index causes a complication of the analysis of resonance positions and prop-
that takes into account the optical properties of inhomogeneous inclusions. Such effective medium approximations have long been used in many fields of physics, especially for the study of the optical and the physical properties of disordered materials.40 Although there has been some criticism concerning the theoretical foundation of the effective medium approximations, the experimental results suggest that some of their forms can be used successfully if the number of inclusions is small, if their size is not too large relative to the wavelength, and if the inclusions are highly absorbing.40 In this case we can expect those properties of a composite inhomogeneous sphere that depend on the bulk material to behave in some respects similarly to the properties of a homogeneous slightly absorbing sphere.

Consequently we have at least some justification to expect an inhomogeneous sphere composed of a small amount of absorbing material dispersed in a dielectric medium to exhibit a resonance structure similar to that of a weakly absorbing homogeneous sphere. However, theoretical arguments can provide only plausibility justification, which should be confirmed or disproved by experiment.

**EXPERIMENTAL RESULTS**

Optical Levitation and Resonances of Homogeneous Dielectric Spheres

We used a linearly polarized argon-ion laser at a wavelength of \( \lambda = 0.5145 \mu \text{m} \) to levitate glycerol droplets (with refractive index \( m = 1.4746 \)) in the radius range between 3 and 10 \( \mu \text{m} \). Part of the radiation of the levitating laser scattered at an angle of approximately 90° is focused onto a silicon photodiode, and the resulting signal is amplified.

**Table 2. Absorbance and Imaginary Part of Re- refractive Index of Three Weakly Absorbing Solutions of Scheaffer Ink in Glycerol**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Absorbance</th>
<th>( m )</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>0.23</td>
<td>2.2 \times 10^{-6}</td>
</tr>
<tr>
<td>S2</td>
<td>0.82</td>
<td>7.7 \times 10^{-6}</td>
</tr>
<tr>
<td>S3</td>
<td>1.15</td>
<td>1.1 \times 10^{-5}</td>
</tr>
</tbody>
</table>

Fig. 2. Example of the measured absorbance of weakly absorbing solutions (S1 and S2) of Scheaffer ink and glycerol as a function of the wavelength.
and fed into a chart recorder. Another part of the radiation scattered at an angle of approximately 90° is incident upon a one-dimensional photodiode array to measure the differential scattering cross section in an approximate angular range 84°-96°. As a droplet slowly evaporates it passes through a set of resonances observed in scattering characteristics whenever the resonance condition for a given partial wave is satisfied. For each resonance peak in the scattering characteristics (in the Mie partial-wave amplitudes \(a\) and \(b\)) there is a corresponding resonance peak in the internal-field-expansion coefficients \(c_n\) and \(d_n\), characterized by a large enhancement of the electromagnetic field close to the surface of a dielectric spherical particle.

The agreement between the measured scattered intensity at 90° and the Mie calculation is excellent, as can be seen in Fig. 1. The scattered intensity was integrated over a solid angle subtended by the collecting lens. Several orders of resonances are observed experimentally. The exact values of width together with the positions of the major resonances between the radii of 4.54 and 4.58 \(\mu\)m are listed in Table 1.

Optical Levitation and Resonances of Homogeneous Weakly Absorbing Spheres

Weakly absorbing liquid was prepared by mixing a small amount of Schaeffer ink (brown no. 1) and glycerol. Mixtures of several different concentrations were prepared, and their spectral transmittance \(T\) near the argon-ion laser wavelength was measured by using a Cary 14 spectrophotometer and \(L = 1\)-cm-long cells. Several examples of the measured spectral absorbance \(A = -\log T\) of the samples are shown in Fig. 2.

The imaginary part of the refractive index of weakly absorbing mixtures at the wavelength of \(\lambda = 0.5145 \mu\)m was calculated from the relation

\[
m_i = -\lambda \ln T/4\pi L,
\]

where \(T\) is the measured transmittance. The imaginary parts of the refractive indices of the four samples used to study the resonance structure of weakly absorbing spheres varied between \(2 \times 10^{-6}\) and \(1 \times 10^{-5}\) (see Table 2 for exact values).

The Mie scattering calculation was done for a set of refractive indices with the same real part of 1.4746 and imaginary parts between \(10^{-7}\) and \(5 \times 10^{-4}\); examples are shown in Fig. 3. The major changes in the resonance structure occur when the imaginary part of the refractive index increases from \(5 \times 10^{-5}\) to \(5 \times 10^{-4}\).

Droplets of each of the mixtures were again levitated by the argon-ion laser radiation pressure, and the resonances in the scattered intensity at an angle of approximately 90° were recorded for the droplet radii between 3 and 10 \(\mu\)m. Examples of the results together with the record of the pure glycerol case are shown in Fig. 4. Within the resolution power of our experimental arrangement the resonance structures of all the cases are identical. This fact is in qualitative agreement with theoretical analysis, since the shift of the resonance positions as well as the reduction of the resonance peak heights should be proportional to the imaginary part of the refractive index, which was in all cases below \(10^{-5}\).
Table 3. Effective Absorbance and Imaginary Part of an Effective Refractive Index of Inhomogeneous Materials*

<table>
<thead>
<tr>
<th>Sample</th>
<th>Grain Size (Diameter in µm)</th>
<th>Absorbance</th>
<th>( m_i ) Corrected for Scattering Losses</th>
</tr>
</thead>
<tbody>
<tr>
<td>H551</td>
<td>0.55</td>
<td>0.20</td>
<td>( 1.88 \times 10^{-6} )</td>
</tr>
<tr>
<td>H552</td>
<td>0.55</td>
<td>0.84</td>
<td>( 7.9 \times 10^{-6} )</td>
</tr>
<tr>
<td>H081</td>
<td>0.08</td>
<td>0.97</td>
<td>( 9.1 \times 10^{-6} )</td>
</tr>
<tr>
<td>H082</td>
<td>0.08</td>
<td>2.05</td>
<td>( 1.9 \times 10^{-5} )</td>
</tr>
<tr>
<td>C1,C2</td>
<td>-0.1</td>
<td>0.09</td>
<td>( 0.85 \times 10^{-6} )</td>
</tr>
</tbody>
</table>

*The materials were composed of hematite particles (H551, H552, H081, and H082) and of carbon (C1 and C2) dispersed in the glycerol host liquid.

Thus we are able to confirm experimentally that the observed resonance structure of weakly absorbing spheres is not changed significantly with the increase in the imaginary part of the refractive index from 0 to \( 2 \times 10^{-6} \).

**Optical Levitation and Resonance Structure of Inhomogeneous Spheres**

To study the resonance structure of inhomogeneous spheres we first used a small amount of hematite particles mixed with glycerol. Two different narrow size distributions of hematite grains (prepared and kindly provided to us in sol form by Milton Kerker and E. Matijevic) were used. The mean sizes of these distributions were 0.55 and 0.08 µm. The refractive index of hematite\(^{29}\) at the 0.5145-µm wavelength is \( m = 3.1 + 0.158i \). Thus we have an inhomogeneous mixture of absorbing hematite particles dispersed in a nonabsorbing glycerol host liquid.

Two different concentrations of each of the size distributions of the hematite particles in glycerol were prepared (altogether four different mixtures). The absorptive property of each mixture is characterized by an effective imaginary part of the refractive index of the mixture. This effective imaginary part of the refractive index is determined from Eq. (1), in which \( T \) is the sample transmittance as determined by using a Cary 14 spectrophotometer. In the case of the hematite–glycerol mixture the deduced imaginary part of the effective refractive index will be overestimated because of the radiation losses inside the cell that are due to the scattering. To correct the imaginary part of the effective refractive index \( m_i \) for the losses resulting from the scattering, we multiplied the results obtained from Eq. (1) by a factor \( Q_{\text{abs}}/(Q_{\text{abs}} + Q_{\text{sc}}) \), where \( Q_{\text{abs}} \) and \( Q_{\text{sc}} \) are, respectively, the absorption and the scattering efficiencies obtained for a spherical hematite particle submerged in glycerol. The correction factor has a value of 0.47 for the large hematite particles (diameter of 0.55 µm) and 0.27 for the small hematite particles (diameter of 0.08 µm) used. The imaginary parts of the effective refractive indices are listed in Table 3.

The resonance structure of the scattered intensity at an angle of approximately 90° of the composite hematite–glycerol droplets within the range of radii from 3 to 10 µm was studied. A small segment of the measurements is shown in Fig. 5. The resonance structure is not changed significantly compared with that of pure glycerol.

Finally, the procedure was repeated with a mixture of black graphitic carbon (soot) submicrometer-sized particles and glycerol. We took \( m = 1.8 + 0.6i \) as a representative refractive index of black carbon, and we calculated \( Q_{\text{abs}} \) and \( Q_{\text{sc}} \) for the spherical carbon particles (of radius \( r = 0.05 \mu m \) in glycerol. The correction factor for scattering losses is \( Q_{\text{abs}}/(Q_{\text{abs}} + Q_{\text{sc}}) = 0.93 \). This value does not change significantly with changes in the refractive index of carbon within the limits of the considered uncertainty of the real (2.0 > \( m_R \) > 1.6) and the imaginary (1.0 > \( m_i \) > 0.4) parts. The effective imaginary part of the mixture determined by using the Cary spectrophotometer (including the correction for scattering losses) is 0.79 \( \times 10^{-6} \) (Table 3). To assure ourselves that the carbon particles are not lost within an atomizer we measured the absorbance of one of our carbon–glycerol mixtures, ran the mixture through the atomizer (used to produce droplets for levitation), collected the droplets produced, and remeasured the absorbance. A comparison of the two absorbance measurements is shown in Fig. 6. Losses of the carbon particles as a result of the process of atomization (droplet production) are not significant.

The resonance structure of the light intensity scattered at an angle of approximately 90° by inhomogeneous spheres...
composed of carbon particles dispersed in glycerol is shown in Fig. 7. The resonance structure itself remains similar to that of a dielectric or a slightly absorbing sphere. However, upon this basic structure we see superimposed erratic random variations in the scattered intensity. We are not sure what causes these apparently random variations in the scattered light. We have verified that it is not the motion of the composite particle itself within the light-levitating trap. The most plausible explanation seems to be the motion of the small black carbon particles within the large composite sphere.

CONCLUSION

From experimental observation we conclude that the resonance structure of weakly absorbing spheres and of slightly inhomogeneous spheres (small mixing ratio of inhomogeneous to homogeneous material) is similar to the resonance structure of dielectric spheres with the same real part of the refractive index. This we have verified for the imaginary part of the refractive index $n_i < 10^{-5}$ (or for the imaginary part of an effective refractive index $n_e < 5 \times 10^{-6}$) and for radii between 3 and 10 $\mu$m.

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REFERENCES


